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Non-monotonic change of electronic properties by As substitution in LaFeP(O,F)

S. Saijo^{a,*}, S. Suzuki^a, S. Miyasaka^{a,b}, S. Tajima^{a,b}

^a Department of Physics, Graduate School of Science, Osaka University, 1-1 Machikaneyama, Toyonaka, Osaka 560-0043, Japan
^b JST, Transformative Research-Project on Iron Pinctides (TRIP), 5 Sanbancho, Chiyoda, Tokyo 102-0075, Japan

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ABSTRACT

The discovery of superconductivity at 28 K in fluorine doped LaFeAsO stimulated a great interest in iron arsenide superconductors. But many differences of physical properties exist between FeP and FeAs 1111 oxypnictide superconductors. To reveal its origin, we synthesized LaFeP_{1-x}As_xO_{0.9}F_{0.1}, and investigated the *x*-dependence of the physical properties. In contrast to a monotonic change of lattice parameters, T_c and T-dependence of resistivity show non-monotonic change.

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1. Introduction

When the superconductivity in LaFeP(O,F) was discovered in 2007 [1], it did not attract much attention because of its low T_c (~5 K). However, after the discovery of higher T_c (~28 K) in LaFeAs(O,F) [2], it has become crucially important to understand the electronic state and the superconductivity mechanism in LaFeP(O,F).

It is a puzzle why the value of T_c in LaFeP(O,F) is so low, compared to that of LaFeAs(O,F). The electronic structures including Fermi surfaces are quite similar in these two systems [3]. The density of states at the Fermi level is expected to be larger in LaFeP(O,F) than that in LaFeAs(O,F) in spite of the lower T_c in the former material. Although the lattice parameters are different between these two materials, it is not understandable that such a small difference in lattice parameters plays an essential role in determining T_c . In the present study, we approach this problem, by investigating the solid solution system LaFeP_{1-x}As_xO_{0.9}F_{0.1} to extract a key ingredient for high T_c superconductivity.

2. Experiments

Polycrystalline samples of LaFeP_{1-x}As_xO_{0.9}F_{0.1} (x = 0, 0.1, 0.2, 0.3, 0.4, 0.6, 0.8, and 1.0) were synthesized by solid state reaction. The mixtures of LaAs, LaP, Fe₂O₃, LaF₃, Fe in the stoichiometric ratio were pressed into pellets in an Ar filled grove box. The pellets were annealed in evacuated quartz tubes at 1100 for 40 h. Here, LaAs (LaP) was obtained by reacting La chips and As pieces (P chips) at 500 (400) for 10 h (15 h), then at 900 (700) for 15 h (10 h).

Corresponding author.
 E-mail address: satsaijo@tsurugi.phys.sci.osaka-u.ac.jp (S. Saijo).

The lattice parameters were determined by powder X-ray diffraction. It was also confirmed that the prepared samples were almost of single phases. Electrical resistivity was measured using a four-probe method.

3. Results and discussion

The lattice parameters a and c were found to increase linearly with increasing As content (x). This proves that a solid solution LaFeP_{1-x}As_xO_{0.9}F_{0.1} was successfully prepared.

Temperature dependences of electrical resistivity are shown in Fig. 1. In all the samples, resistivity shows a sharp superconducting transition, which enables us to determine T_c from zero resistivity temperatures. The T_c values of both end materials were identical to the previously reported ones [1,2]. As we see in the upper panel of Fig. 2, T_c gradually increases with x up to 0.6, while it is saturated or weakly decreased above x = 0.6.

In contrast to the monotonic change in lattice parameters, resistivity changes with x in a complicated manner, as shown in Fig. 1. We can discuss the resistivity change from the three viewpoints. The first is power of T at low temperatures, the second is the residual resistivity, and the third is the slope at high temperatures.

T-dependence of resistivity can be expressed as $\rho(T) = \rho_0 + AT^n$. For x = 0, n is close to 2, indicating that the x = 0 end material is a conventional Fermi liquid [4]. When x exceeds 0.2, the power (n) rapidly decreases and reaches about unity at x = 0.6. Above x = 0.6, n increases again and becomes close to 2. (See the lower panel of Fig. 2.) The *T*-linear resistivity down to 30 K is similar to the behavior of high- T_c cuprates, which strongly suggests that the conduction mechanism is governed by a strong spin fluctuation [5]. The x dependence of n indicates that the antiferromagnetic fluctuation rapidly increases with x up to x = 0.6, followed by a weak decrease above 0.6. At x = 1.0, antiferromagnetic fluctuation





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Fig. 1. Temperature dependence of resistivity for $LaFeP_{1-x}As_xO_{0.9}F_{0.1}$ with various xs.



Fig. 2. As content (*x*) dependence of T_c (upper) and power of *T* in $\rho(T)$ (lower).

is stronger than that for x = 0, although it is strongest not in the end material (x = 1.0) but at $x \sim 0.6$.

Since the present samples are polycrystals, it is hard to discuss the absolute value of resistivity. But we can see a rough tendency in Fig. 1 that residual resistivity increases with *x*. There seems to be a jump between x = 0.2 and 0.4. Partial replacement of P by As may introduce scattering centers in a conduction path. However, a larger residual resistivity even in the end material LaFeAs(O,F) cannot be understood in terms of the disorder effect. There seems no disorder effect on T_c , because T_c increases with introducing P in LaFeAs(O.F).

The slope of $\rho(T)$ at high temperatures also shows a distinct behavior in the intermediate composition region (0.4 < x < 0.8). The slope increases with *x* and reaches the maximum value at $x \sim 0.8$, followed by a small decrease at x = 1.0. In a conventional metal, the high *T* slope of resistivity is proportional to the electron–phonon coupling strength. In the present case, since spin fluctuation dominates conduction mechanism, the slope of $\rho(T)$ may reflect other quantities. However, a rough tendency that the slope is the largest near x = 0.8, which correlates with *n* and ρ_0 , is very suggestive in relation to the electronic state basing on superconductivity.

From the common *x*-dependences of all the three parameters as well as T_c , we conclude that there is some critical change between x = 0.2 and 0.4. Judging from the *T*-linear behavior of resistivity, we can expect that the antiferromagnetic fluctuation is stronger in this intermediate region. This may be linked to a rapid increase in T_c in this region.

The presence of a quantum critical point was suggested in the $BaFe_2(As,P)_2$ system, based on a rapid change in *n*-value [6]. The present result that T_c starts to increase when *n* becomes close to unity is similar to the situation in $BaFe_2(As,P)_2$. However, in the present case, both of the two phases are a superconducting phase, while in the case of Ref. [6], a SDW phase is competing with a superconducting phase. The present result indicates that two superconducting phases are separated by some intermediate phase where antiferromagnetic fluctuation is very strong. In order to clarify the meaning of the observed unusual electronic change, further studies in both experiments and theories are necessary.

4. Summary

The solid solution system of two superconductors LaFePO_{0.9}F_{0.1} and LaFeAsO_{0.9}F_{0.1} was successfully prepared over a whole range of composition of As/P. In contrast to the *x*-linear changes of lattice parameters, T_c shows a non-monotonic change. When a rapid increase in T_c is observed at $x \sim 0.3$, the *T*-dependence of resistivity changes from T^2 -behavior to *T*-linear one. Since the latter behavior is an indication of strong spin fluctuation, the enhancement of spin fluctuation can be considered to be linked to a rapid raise of T_c near x = 0.3.

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